Beamline 1.4.2

Visible and Infrared Fourier Transform Spectroscopy (FTIR)

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Absolute Flux Measurements for IR Spectromicroscopy

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1. INTRODUCTION

Measurements of the effects of exposure to IR light at the spectromicroscope at beamline 1.4.3 has necessitated an estimate of the absolute flux levels in the IR optical bench and in the microscope. We calculated the IR flux levels from both a thermal source and the ALS, and compared them to measurements taken at the beamline. We find a self consistent picture which can be used to estimate the absolute exposure to cells and bacteria which are examined in the IR microscope.

2. EXPERIMENTAL

The thermal ("globar") source in the Nicolet 760 IR bench was measured with a disappearing filament style optical pyrometer to have a brightness temperature of ~1343K. This allowed us to calculate the "greybody power" emitted by the filament, using the emissivity of tungsten. We also measured the output of the thermal source with a Molectron PowerMax 500D with a 19 mm dia. PM10V1 detector. To measure ratios of light both before and after the bench and the microscope a more sensitive mid IR detector was needed so we employed a T3-09 9mm dia. energy detector, and a 200 Hz chopper. We also calculated the power of the synchrotron beam using the standard bending magnet formulae. The table below summarizes our results. Unless enumerated otherwise the numbers are in watts. Bold numbers are experimental data, others are calculations or estimates.

Greybody power	1.70E-04	**0.0479**	Synchrotron Power	3.73E-02
1 to 20 microns			1 to 20 microns	
1 mm^2, 0.0064 str, 1343 K			10 x 40 mr, 1.9 GeV, 4.81 m	
Real emitting area, mm^2	100	1	Beamline coupling loss	0.333
			20 out of 40 mr * 0.67 diamond	
Bench efficiency	0.372	0.372	Bench efficiency	0.372
Chopped Energy Detector			Chopped Energy Detector	
15.9/42.7			**15.9/42.7**	
Microscope Vignetting	0.1677	0.1677	Microscope Vignetting	1
Chopped Energy Detector				
1.5/15.9 * 12^2/9^2				
Microscope efficiency	0.225	0.225	Microscope efficiency	0.225
0.6/(1.5*12^2/9^2)			**0.6/(1.5*12^2/9^2)**	
Chopped Energy Detector			Chopped Energy Detector	
Power at sample	2.39E-04	6.72E-04	Power at Sample	1.04E-03
Greybody			Synchrotron	
Ratio of Synch/Greybody	**2**		Peak Power at Sample	5.20E-02
by MCT in Microscope			Synchrotron (*50)	
Ratio from this work	1.55		for 2ns/40ps	

3. RESULTS AND DISCUSSION

Starting from two calculations, and one measurement the table precedes from top to bottom from the two sources, thermal and synchrotron to the sample. At each step, there were many factors to consider. For example, the long depth of field of the synchrotron source means that all 40 horizontal mr of beam is not coupled into the microscope, and the synchrotron beam has a diamond window loss which the thermal beam does not. Moreover, the synchrotron beam has been carefully moved from the center of the optical axis so that it is not vignetted by the special coating in the center of the beam splitter in the optical bench, and the microscope optics have been adjusted to optimally transmit the synchrotron beam. This requires that the vignetting ratios for the two cases be different. We have done our best to honestly estimate the various factors, and measure them where feasible without significant disassembly of the beamline optics.

The final results are satisfying in that the ratio of the total synchrotron to thermal flux (1.55) that we get from our careful combination of theoretical and experimental measurements matches well within the error of our methods the ratio (2.0) we measure by comparing the two sources directly with the LN_2-cooled MCT detector in the microscope. The brightness advantage of the synchrotron (>100) is also verified. The overall error in the estimates is well represented by the factor of 2.8 between the two methods for estimating the power of the thermal source. The continuous power at the microscope integrated from 400 to 10000 cm^{-1} is estimated to be approximately 1 mW, and the peak power at the top of an ALS pulse to be fifty times (2 nsec / 40 psec) that, or 50 mW.

ACKNOWLEDGEMENTS

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Coherent Far IR Bursts Measured at BL 1.4.2

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INTRODUCTION

We have performed the first measurements at the ALS of coherent far-IR bursts coming from instabilities within a high-current single electron bunch.

The 2-bunch mode operations of the ALS afforded us the opportunity to use the machine while it was tuned for high currents in individual electron bunches. We have used a number of accelerator physics shifts to make measurements with well-controlled setups as well as additional measurements during normal 2-bunch operations. Recent investigations at other light sources [1-6] have observed far-IR bursting at high bunch currents. We want to investigate these bursts more carefully towards the goal of understanding how to make use of high-intensity coherent far-IR synchrotron light as a new source of far-IR that is many orders of magnitude brighter than the best presently available sources.

TIME DOMAIN MEASUREMENTS

As an initial investigation into coherent far-infrared synchrotron radiation we placed a liquid He cooled Silicon Bolometer with integrated pre-amplifiers just outside a 20 mm diameter diamond window mounted in the 'switchyard' at Beamline 1.4. A single extra mirror was inserted in the switchyard to direct the collimated beam through this window into the bolometer without disturbing the alignment of the IR beamlines. A digitizing oscilloscope recorded the output of the detector. We observed large intensity bursts when the single bunch current was very high. Fig. 1 shows time traces of the bolometer output voltage for three different beam currents in single bunch operation. Although the bursts seemed to be quasi-random, at certain currents the bursts occurred within a periodic envelope, as evidenced by the middle trace in Fig. 1. The rise and fall times of the bursts were detector limited, therefore the damping mechanism may not be inferred immediately. The single bunch current threshold for the onset of

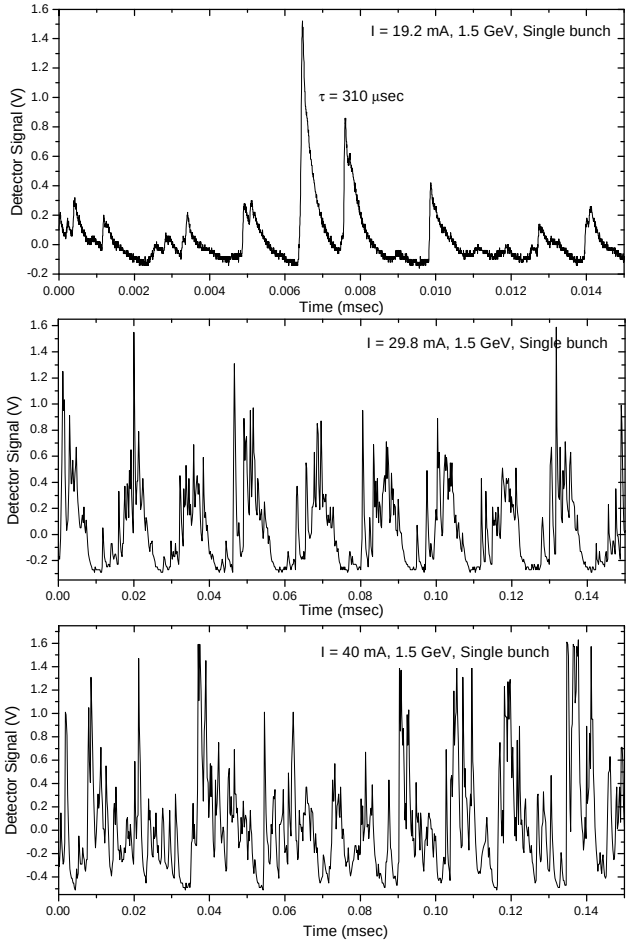

Figure 1. Far-IR detector signal vs time for three different beam currents. Note the expanded time scale for the top panel.

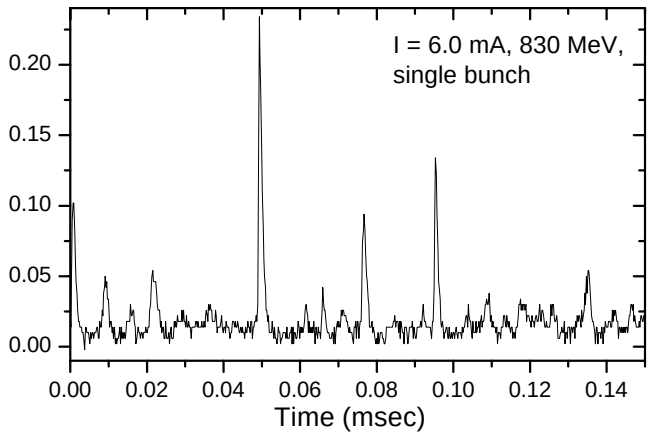

Figure 2. Bursting observed at 830 MeV which shows a quite different time envelope.

bursts is approximately 7 mA at 1.5 GeV. The transition to bunching of the bursts within a super period occurs at about 27 mA at 1.5 GeV.

Since the bursts seem to be related to high peak currents within a bunch, reducing the RF power will lengthen the bunch and therefore cause the bursts to stop. At 14 mA and 1.9 GeV, the bursts go away by reducing the RF power from 120 to 90 KW; returning to 120 KW causes the bursts to reappear. At 1.5 GeV and 14 mA, the bursts can be stopped by dropping the RF power from 120 to 1.5 KW.

A measurement of the bursting at 830 MeV electron energy at 6 mA single bunch current showed distinctly different time behavior as shown in Fig. 2.

SPECTRAL MEASUREMENTS

By using a combination of filters with the above setup we determined the spectral content of the bursts was below 100 cm^{-1} (wavelengths longer than 100 microns). To make this more quantitative, we used the Bruker 66v/S FTIR spectrometer on BL 1.4.2 to measure the bursts that occur at the very top of the fill during regular 2-bunch mode operations. By quickly averaging 100 interferograms over 50 seconds we integrated long enough to demonstrate the average spectral content of the bursts. Shown in Fig. 3 is the measured intensity of the bursts as a function of wavelength, ratioed to the incoherent synchrotron signal measured at low beam current. The bursts are peaked at ~27 cm^{-1}. This indicates a microbunching within the electron bunch having a period on the order of 400 microns (or approximately 30 times smaller than the normal ALS bunch length). The bursting was clearly dependent on the beam energy, with higher intensity at 1.5 GeV. This is to be expected, as the bunch length is proportional to $E^{3/2}$, implying higher peak currents at 1.5 GeV than 1.9 GeV, and therefore a greater tendency for instabilities and hence microbunching. Overall, the burst intensity dropped with decreasing beam current, with the spectral content remaining essentially unchanged.

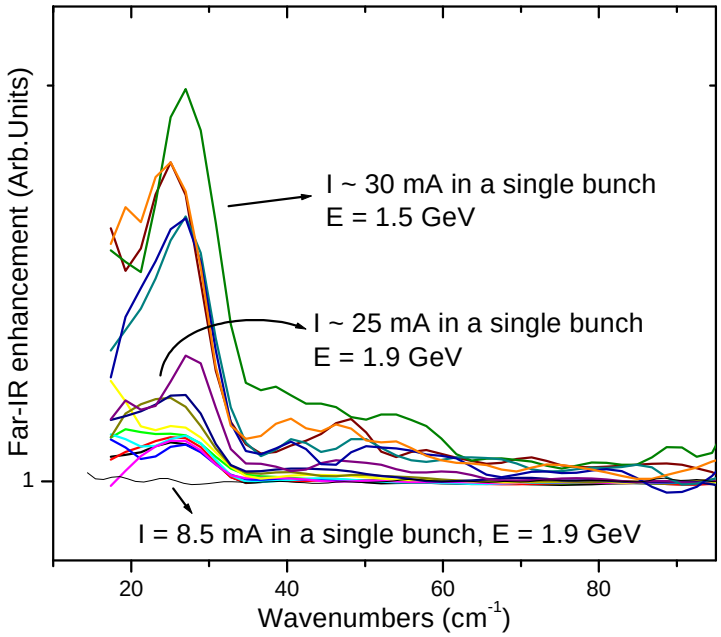

Figure 3. Spectrum of coherent far-IR bursts measured at Bl1.4.2 as the current was decaying.

We are continuing these measurements to gain a greater understanding into coherent synchrotron emission both

from instabilities (as measured here) and hopefully in a stable manner. In the near future, we will make additional measurements using femtosecond sliced electron bunches (in collaboration with the femtosecond x-ray team at LBNL) which will allow a clean study of coherent emission from a well known transient short bunch. Because this will be a laser pumped measurement with synchronized optical detection, electron beam instabilities and oscillations will not be a factor. We have also collaborated with researchers at Brookhaven National Laboratory and Thomas Jefferson National Laboratory to measure the coherent far-IR emitted from a bend magnet in their energy-recovery linear accelerator based infrared free electron laser [7] (reported in a separate compendium abstract).

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Giant Terahertz Power Levels from Relativistic Electrons

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INTRODUCTION

We present measurements of both power and spectral content that confirm theoretical predictions of the generation of 20 watts (average) of broadband THz radiation pulses. The experiments were performed using the energy recovery linac (ERL) at the Jefferson Lab Free Electron Laser (JLab FEL)[1]. This facility offers a combination of very short electron bunches (~ 500 fs), relatively high average beam current (up to 5 ma), and up to a 75 MHz repetition rate.

The THz region, (1 THz = 33 cm^{-1} or 4 meV), lies in the far infrared spectral range where conventional thermal sources are very weak. A significant advancement in broadband THz sources has occurred over the past decade with the advent of coherent THz radiation emission from photocarriers in a biased semiconductor[5]. An energy per pulse of about 1μJ has been achieved, implying MW peak powers, but at repetition rates of 1 kHz such that average power levels are only 1 mW[5].

The present work describes a different process for producing coherent THz radiation by accelerated electrons. As schematically shown in Figure 1, electrons are photoemitted from GaAs, brought to relativistic energies (~ 25 MeV) in a linac and then transversely accelerated by a magnetic field to produce the desired THz emission as synchrotron radiation. If the electron bunch dimensions are small (in particular, the bunch length is less than the wavelength of observation), one obtains a multiparticle coherent enhancement[6,7].

Conceptually it is easy to understand the many orders of magnitude gain realized in these experiments by making a comparison with a more conventional (non-relativistic) THz source. We can compare the power produced per electron, and use Larmor's formula[19] for the radiated power. In CGS units it takes the form:

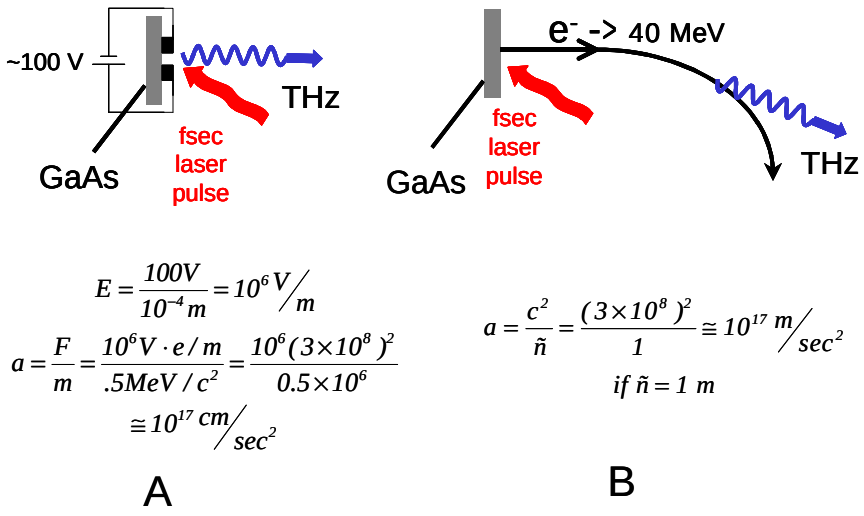

Fig. 1. Comparison between coherent THz radiation generated by a conventional laser-driven THz source (A) and the relativistic source described here (B).

$$Power = \frac{2e^2a^2}{3c^3}g^4$$

where e is the charge, a is the acceleration, c the speed of light and γ is the ratio of the mass of the electron to its rest mass. In case A (the conventional THz emitter), these carriers immediately experience a force from the bias field (~100 volts across a 100 micron gap) of ~10^6 V/m, which results in an acceleration of

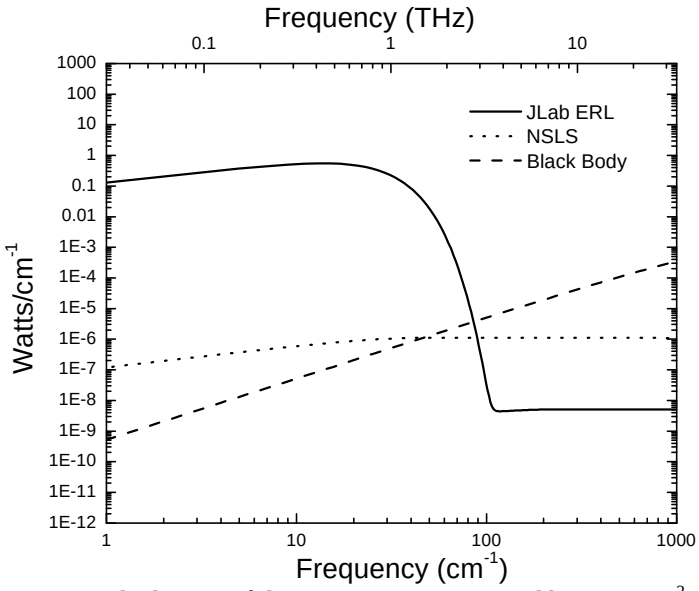

Fig. 2. Calculations of the average power emitted by a 10 mm^2 2000 K thermal source (dashed), the NSLS VUV ring at BNL (dotted), and the JLab ERL (solid).

10^{17} m/s^2. The entire process is completed in less than 1 ps, resulting in spectral content up to a few THz. In case B, the same number of charge carriers are brought to a relativistic energy of > 10 MeV in a linac, after which a magnetic field bends their path into a circle of radius $r = 1$m resulting in an acceleration $c^2/r = 10^{17}$ m/sec^2, the same as for case A. An observer for case B also detects a brief pulse of electromagnetic radiation as an electron bunch passes by. The bunch length determines the spectral range over which the coherent enhancement occurs. For an electron energy of 10 MeV ($\gamma = 21$), and with $r = 1$ m, we obtain a dt of about 500 fs, which is comparable to the bunch length. The resulting spectral content extends up to about 1 THz, the same spectral range as for case A. Thus, assuming the same number of electrons, the ratio of the power radiated by the conventional THz generation to the present generation is given by $\gamma^4 = 2 \times 10^5$, with $\gamma = 21$. In practice, the electron energy can be significantly larger, but this simply adds relatively weak (incoherent) intensity at higher frequencies and leaves the low frequency (THz) intensity essentially unchanged.

The results of calculations of the radiation for a conventionally synchrotron, a thermal IR source, and the JLab source are shown in Fig. 2 in units of (average) W/cm^{-1} over the range 1-1,000 cm^{-1}, or 0.03 to 33 THz. The superiority of the JLab ERL in the THz range is clear.

EXPERIMENTAL RESULTS AND DISCUSSION

In our experiments, the electrons were generated using the frequency doubled (530 nm) output of a Coherent Antares model Nd:YLF laser operating at sub-multiples of frequencies up to 74.8 MHz, and with an average power of a few watts incident on a Cs coated GaAs cathode. The resulting photoelectrons were accelerated using a DC voltage of 300 kV into a superconducting linac and accelerated to energies of up to 40 MeV.

The THz radiation was extracted from a dipole magnet of 1 m bending radius immediately prior to the free-electron laser cavity, the latter being unimportant for these experiments. For the total power measurements the light exited the accelerator vacuum chamber through a 10 mm aperture diamond window subtending an angle of 20 × 20 mrad relative to the source point. The emerging beam was focused onto a calibrated pyroelectric detector, with which the total power was measured, and which confirmed the predictions of the calculations for this aperture, charge per bunch and repetition.

The spectral content of the emitted THz light was analyzed using a Nicolet 670 rapid-scan Michelson interferometer with a silicon beamsplitter and a 4.2K Infrared Laboratories bolometer. The diamond window on the accelerator was replaced by a larger crystal quartz window to increase the light collection to 60 × 60 mrad. An 80 cm focal length spherical mirror produced a 48 mm diameter collimated beam compatible with the Nicolet 670 interferometer optics. A

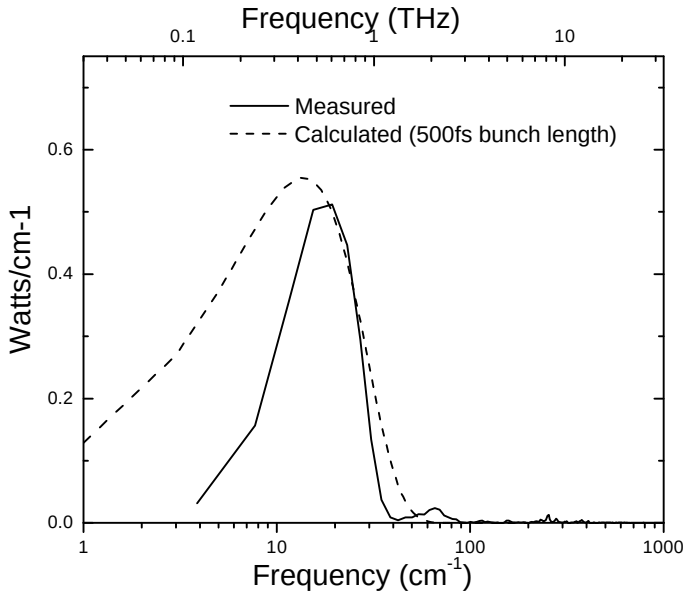

Fig. 3. Comparison between measured (solid line) and calculated (dashed line) THz spectral intensity.

switching mirror allowed a remote choice of source, namely the THz light from the accelerator, or a T=1300 K thermal reference source.

For the spectroscopy experiments, the analysis and detection system did not have sufficient dynamic range to cover the 7 decades in power difference between the 2 sources. We chose to make measurements at 584 kHz, instead of 37.4 MHz, and at a charge per bunch of 34 pC instead of the maximum of 100 pC, thereby reducing the THz power by a factor of $(37 \times 10^6) / (584 \times 10^3) \times (100/34)^2$, or approximately 550.

We show the results of the measurements along with a calculated curve for a bunch length of 500 fs in Fig. 3. We have scaled the data to fit the theory on the basis of the absolute power measurements. The spectral onset of the super-radiant enhancement is clearly seen, and the onset shape is also seen to match closely the theoretical predictions. Note that the discrepancy on the lower frequency side is due to diffraction effects.

We have another reference point for determining the absolute power since we were able to switch sources from the THz emission port to a 1300 K thermal source (the spectrometer's standard globar source). At a frequency of 12 cm^{-1} we obtained a ratio of intensity from the THz source to that of the globar of 2×10^4. To compare with the calculation, we multiply the THz source results by the reduction factor of 550, as discussed earlier. This implies a measured advantage of the JLab THz source over the globar of 10^7. The calculation predicts an enhancement of $(0.6 / 6 \times 10^{-8}) = 10^7$. While there is apparent agreement, these simple arguments have ignored diffraction and other effects on the detection efficiency of both sources. However, the result does indeed affirm the THz power.

We additionally observed the expected quadratic dependence of the coherent THz intensity on the number of electrons in the bunch, and the intensity ratio for the horizontal to vertical polarization components was measured to be 5. The expected polarization ratio for 30 cm^{-1} and 60 mrad is about 6, so we consider this to be good agreement.

CONCLUSIONS

We have demonstrated that the short bunches which circulate in energy recovery electron circulating rings with sub-picosecond electron bunch lengths yield broadband high brightness THz radiation with close to 1 W/cm^{-1} of average power into the diffraction limit and peak powers about 10^4 times higher than this.

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Infrared conductivity of photocarriers in organic molecular crystals

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This project, begun in March of 2001, seeks use infrared spectroscopy to probe the properties of photocarriers in organic molecular crystals. A group at Bell Laboratories has recently succeeded in creating FETs on ultrapure single crystals of pentacene, tetracene, and other "polyacenes" (rigid, rodlike chains of Benzene rings). Carriers injected into these transistors have exhibited metal-insulator transitions, superconductivity, lasing, and the quantum Hall effect (see, e.g., J. H. Schoen, Ch. Kloc, B. Batlogg, *Science* v. 288 p. 2338; *Science* v. 288 p. 656; *Nature* v. 406 p. 702). The goal of our experiment is to photoexcite carriers in these same materials, and to measure the infrared spectrum of these photocarriers.

The apparatus, at ALS beamline 1.4, includes an Argon-ion laser for photoexcitation of charge carriers, a Bruker 66v/S Fourrier-Transform Infrared (FTIR) spectrometer, and a variable-temperature cryostat. The laser allows photoexcitation at energies from 2.4 eV to 3.5 eV. At the UV frequencies, the quantum efficiency of photocarrier generation in pentacene is high, about 30%. The light is coupled to the sample (in the cryostat) by means of an optical fiber. The cryostat allows us to reach temperatures from 5 K to 300 K, and the FTIR spectrometer allows measurement of infrared transmission on the range of at least 100 cm^{-1} to 7800 cm^{-1}.

The basic result of a measurement is a transmission spectrum, $T(\omega)$, either with the sample illuminated with laser light, $T_{on}(\omega)$, or with the sample unilluminared, $T_{off}(\omega)$. From these spectra we determine difference spectra, $\Delta T / T = (T_{on}(\omega) - T_{off}(\omega)) / T_{off}(\omega)$. Our measurements at room temperature on crystals of Tetracene have shown good reproducibility, both in the shape and in the magnitude of the difference spectra. The measurement is sensitive to changes in the transmission of one part in 10^4 or better through most of the spectral range. We have resolved many clear and reproducible features that are an order of magnitude larger than the noise.

Although these results demonstrate that the apparatus is, indeed, able to make the desired measurements, they do not put us particularly close to our scientific goals. Most or all of the features we have seen thus far appear to be due to shifting or broadening of phonon absorptions due to the heating effect of the laser. This result at room temperature is no surprise, as the mobility of carriers in polyacene crystals increases as T^{-2} below room temperature, so that the conduction of the carriers should only become visible at low temperatures. More surprising was the result that tetracene crystals large enough to measure optically will invariably shatter upon cooling below about 180 K (the temperature seems to vary a bit). Pentacene crystals, on the other hand, do not shatter, but also do not seem to be available in sizes large enough for optical measurement. Other

polyacenes have much lower efficiencies of photocarrier generation (due to their having broader bandgaps), and so are not suited to this experiment.

We are now seeking thin-film samples of the polyacenes, as these should survive cooling and should have the large area desirable for infrared measurement. High-quality pentacene films have been shown to display many of the same electronic properties as their single-crystal counterparts (J. H. Schoen, Ch. Kloc, *Applied Physics Letters*, v. 79, p. 4043).

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New IR microscope and bench installed at BL1.4

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1. INTRODUCTION

New infrared spectromicroscopy equipment was purchased for and installed on the ALS infrared beamlines on beam port 1.4. It includes the latest step-scan capable FTIR bench and an infinity corrected infrared microscope which will allow for a number of new sample visualization methods. This equipment was purchased with funding from the DOE Office of Biological and Environmental Research (OBER) with the express purpose to develop biomedical and biological applications of synchrotron-based infrared spectromicroscopy.

2. EQUIPMENT

The new spectromicroscopy equipment includes a Thermo Nicolet Nexus 870 step- and rapid-scan FTIR bench, and a Thermo Spectra-Tech Continuum IR microscope, photographed below. The IR microscope includes two IR detectors, a wide-band MCT and a fast (20 ns) TRS MCT for time-resolved experiments. A fast digitizer (up to 100MHz) compliments the TRS MCT detector. The synchrotron beam coupled into the IR microscope continues to have a diffraction-limited spot size, thereby attaining a 200-fold increase in signal from small (3 – 10 micron) sample spot compared to a conventional thermal IR source. The infinity-corrected microscope optics allow for a number of additional sample visualization accessories which can help the user identify the important location within their sample for micro-IR analysis:

- Visual and IR polarizers
- Dark-field illumination
- DIC (Differential Interference Contrast) optics
- UV Fluorescence

An example of DIC optics enhancing a micrograph of human cheek cells is shown in the photograph to the right. The DIC technique provides a psuedo-3D effect, enhancing the contrast between different thicknesses of an otherwise clear sample. In the image to the right, one can make out the nuclei of the cells (thicker bump near the middle of each cell), whereas this would be difficult using conventional illumination.

This new instrument will aide in user scientific research across many fields. For example, the study of individual living cells, toxic contaminants, bioremediation, protein microcrystals, rhizoids, and forensic evidence will all be enhanced by the additional capabilities of this new SR-FTIR spectromicroscopy system.

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